

IMPACT OF ANTHROPOGENIC SULFUR EMISSIONS ON CLOUD-CLIMATE INTERACTIONS

Catherine C. Chuang and Joyce E. Penner

Lawrence Livermore National Laboratory
Livermore, CA 94551, U. S. A.

1. INTRODUCTION

It has been estimated that anthropogenic sulfur emissions dominate over natural sulfur emissions by about a factor of 2 on a global average (Spiro et al., 1992) and they have been significantly increased during the past several decades (Charlson et al., 1992). Photochemical reactions of the emitted anthropogenic sulfur compounds lead to a large increase in the number concentration of sulfate-containing aerosols. These soluble aerosols modify the microphysics of clouds by acting as cloud condensation nuclei (CCN) and enhancing the reflectivity of low-level water clouds. They might also alter the development of precipitation by affecting the mean drop size and thereby influencing cloud lifetime. However, the magnitude of cloud optical property change associated with anthropogenic sulfur emissions is quite uncertain because of the large uncertainty in the relationship of aerosol size distribution to sulfate mass concentration. There are further complications as a result of the non-linearity of cloud microphysics where the cloud drop number concentration is a function of updraft velocity and aerosol characteristics. In this paper, we coupled a climate model with a 3-D chemistry model to simulate the global sulfur cycle and estimated radiative forcing by anthropogenic sulfur emissions through cloud processes (i.e., indirect forcing).

2. FORMATION OF PARTICULATE SULFATE

To assess the impact of anthropogenic sulfate on cloud optical properties, two different approaches for the formation of anthropogenic sulfate are applied in this study. The first one is referred to as the internal mixing approach where aerosols with the same size are assumed to have identical chemical composition. The aerosol size distribution is thus determined by the condensation of sulfuric acid vapor (H_2SO_4) on a prescribed pre-existing particle distribution and by aqueous-phase oxidation of SO_2 followed by evaporation of cloud drops. Langner and Rodhe (1991) estimated that the amount of sulfate transformed by OH oxidation is between 19% and

55% of that by in-cloud oxidation. Thus, if sulfate is produced by these two pathways only, 65% - 85% of sulfate would be transformed by in-cloud oxidation of SO_2 . These lower and upper values are used in this work to provide an estimate of the sensitivity of the predicted change of cloud drop number concentration to the assumed fraction of different oxidation pathways. We simulate the aerosol size distribution using the process described by Chuang and Penner (1995) with a prescribed pre-existing particle distribution. The continental and marine aerosol size distributions simulated by the internal mixing approach with the fraction of anthropogenic sulfate converted through the aqueous pathway specified as 85%, 75%, and 65%, respectively, are shown in Fig. 1.

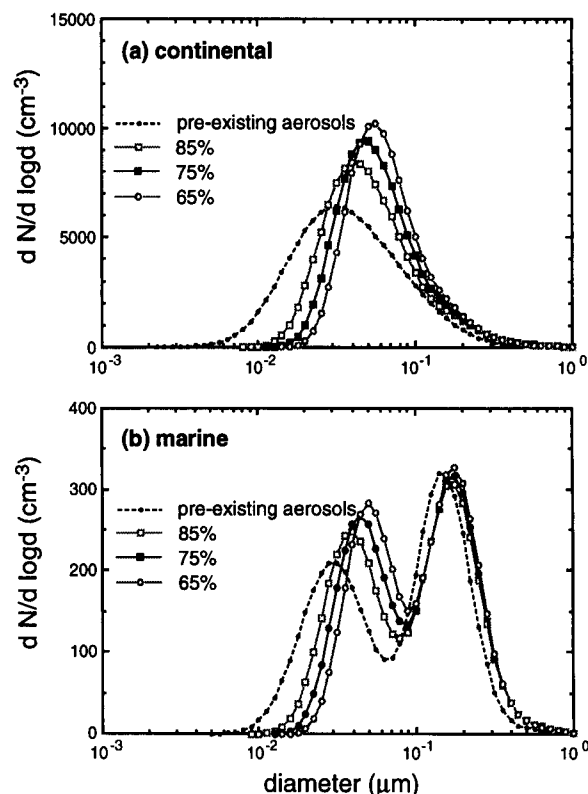


Fig. 1. Simulated aerosol size distributions. Aerosol number and anthropogenic sulfate mass concentrations are 5500 cm^{-3} and $5 \mu\text{gm}^{-3}$ for continental case; 250 cm^{-3} and $0.5 \mu\text{gm}^{-3}$ for marine case.

The second approach is referred to as external mixing where the particulate sulfate is formed by homogeneous nucleation or condensation onto pure existing sulfate particles. Pure sulfate might also be formed in a drop which formed on a pure pre-existing sulfate particle. In this approach, aerosols with the same size would have different chemical composition. The anthropogenic sulfate aerosols are assumed to have a size distribution from fitting the average measured sulfate data summarized by Milford and Davidson (1987). The fitted sulfate aerosol mass distributions as well as their corresponding number distributions in the external mixing approach are presented in Figs. 2a and 2b. Since the sulfate data measured in the marine boundary layer contains sea spray aerosols, the coarse-particle sulfate is attributed to sea salt. Therefore, we only fit the measured data with particle size $< 1 \mu\text{m}$ for the marine case.

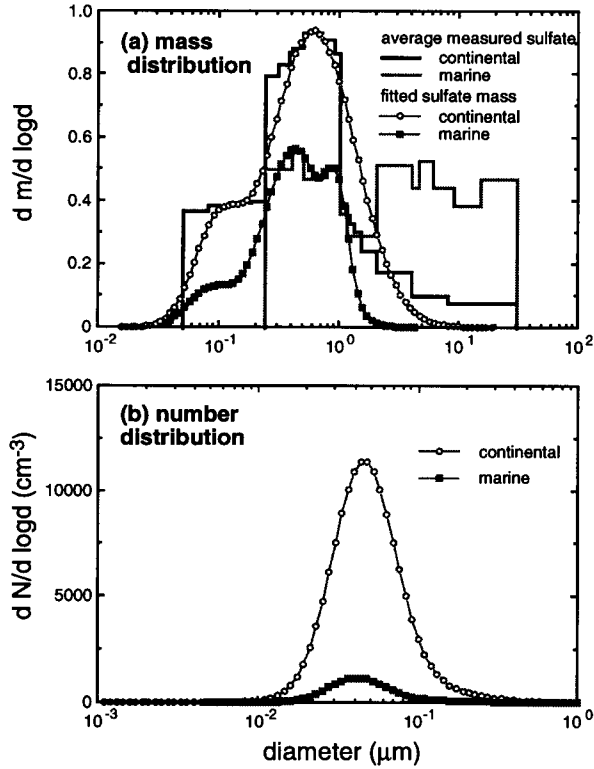


Fig. 2. (a) Dimensionless sulfate mass distributions. (b) Fitted sulfate aerosol number distributions. The number concentration is 5960 cm^{-3} for the continental case and 580 cm^{-3} for the marine case. These values correspond to anthropogenic sulfate concentrations of $5 \mu\text{g m}^{-3}$ and $0.5 \mu\text{g m}^{-3}$, respectively.

Our coupled climate/chemistry model together with detailed emission inventories is used to predict the global aerosol distribution. Figure 3 presents the simulated seasonal variation of aerosol number concentration. Figure 3a shows that the lowest global average aerosol number concentration is in April, a time in between two high seasons of aerosol generation by biomass burning in winter and summer.

Figures 3b and 3c reveal that anthropogenic sulfate reaches its peak magnitude during the N. Hemisphere summer time. It is also noted that biomass burning is the major aerosol source in the S. Hemisphere.

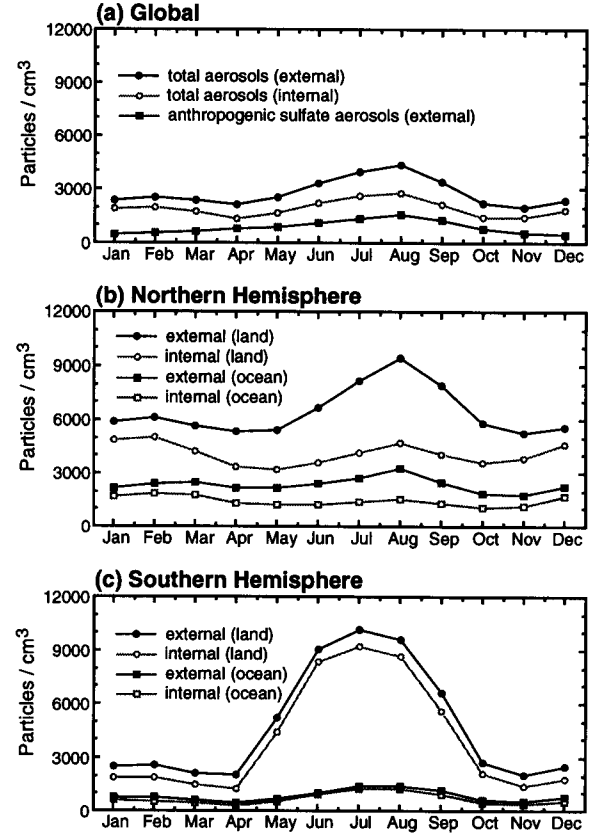


Fig. 3. Simulated seasonal variation of aerosol number concentration.

3. CLOUD DROP PARAMETERIZATION

A series calculations have been performed to investigate the relationship between aerosol size distribution and cloud drop number concentration. We parameterized the number concentration of cloud drops activated through the nucleation process using a detailed microphysical Lagrangian model (Chuang et al., 1992). Following Chuang and Penner (1995), the drop number concentration (in cm^{-3}) is expressed in the form of $N_d = w N_a / (w + c N_a)$, where N_a (cm^{-3}) is the aerosol number concentration, and w (cm s^{-1}) is the updraft velocity. The coefficients c developed from two different approaches are listed as follows

(a) Internal Mixing Approach

$$\text{over land } c = 0.0410 + 21.59 X_L$$

$$\text{over ocean } c = 0.0222 - 0.133 X_O + 3.074 X_O^2$$

where $X_L = \log w [1 - \log w (.5 + \gamma/\alpha^4) / (\log N_a)^2] / (\log N_a)^{5+\gamma/\alpha^3}$, and $X_O = \log w [1 - \log w (.5 + .2\gamma/\alpha^3) / (\log N_a)^2] / (\log N_a)^{2+1\gamma/\alpha^2}$. γ is defined as the ratio of anthropogenic sulfate loading (in $\mu\text{g m}^{-3}$) to the total aerosol number (in 1000 cm^{-3}). α is the ratio of the

fraction of anthropogenic sulfate converted by the aqueous pathway to the mean value used here (75%).

(b) External Mixing Approach

$$\text{over land } c_s = 0.0412 + 3.698 X_{s,L}$$

$$c_b = 0.0410 + 21.59 X_{b,L}$$

$$\text{over ocean } c_s = 0.0117 + 5.015 X_{s,O}^2$$

$$c_b = 0.0222 - 0.133 X_{b,O} + 3.074 X_{b,O}^2$$

where c_s and c_b are coefficients for anthropogenic sulfate aerosols and pre-existing particles with number concentrations N_s and N_b , respectively. $X_{s,L} = \log w [1 - .055 (\log w)^2 \log N_s] / \log N_s^4$, $X_{b,L} = \log w [1 - .5 \log w / (\log N_b)^2] / (\log N_b)^5$, $X_{s,O} = \log w [1 - .525 \log w / (\log N_s)^5] / (\log N_s)^{3.5}$, and $X_{b,O} = \log w [1 - .5 \log w / (\log N_b)^2] / (\log N_b)^2$. The total aerosol number concentration $N_a = N_s + N_b$, and cloud drop number concentration nucleated is approximated by $N_d = (N_s/N_a)w N_a / (w + c_s N_a) + (N_b/N_a)w N_a / (w + c_b N_a)$.

These parameterizations are introduced into the coupled climate/chemistry model to predict the cloud drop number concentration. Simulated seasonal variation of the percentage change in cloud drop number concentration due to anthropogenic sulfate is shown in Fig. 4. Both approaches indicate that April has the highest percentage increase in cloud drop concentration.

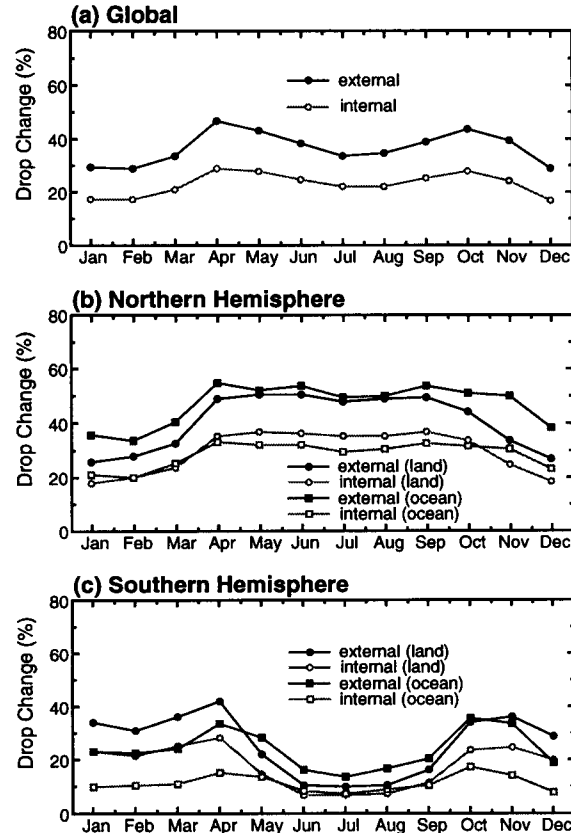


Fig. 4. Simulated seasonal variation of the percentage change in cloud drop number concentration due to anthropogenic sulfate.

4. INDIRECT ANTHROPOGENIC SULFATE RADIATIVE FORCING

We calculate the indirect anthropogenic sulfate radiative forcing as the difference in top of the atmosphere solar radiation with and without anthropogenic sulfate at each time the radiation routine is called. Results are listed in Table 1.

Table 1. Global average annual mean indirect anthropogenic sulfate forcing (Wm^{-2})

	Internal mixing with aqueous conversion fraction			External mixing
	65%	75%	85%	
N. H.	-1.74	-1.25	-0.91	-2.15
Land	-1.37	-0.99	-0.73	-1.55
Ocean	-1.98	-1.41	-1.02	-2.53
S. H.	-0.75	-0.49	-0.34	-1.12
Land	-0.64	-0.44	-0.31	-0.76
Ocean	-0.77	-0.51	-0.35	-1.20
Global	-1.24	-0.87	-0.62	-1.63

The indirect effect is much more pronounced over the ocean where the pre-existing aerosol number concentrations are lower than over continental areas. This low concentration yields a higher fractional increase in cloud drop number for the same amount of anthropogenic sulfate deposited (Chuang and Penner, 1995). Because of the significance of the indirect effect over the ocean, we investigated the sensitivity of the indirect forcing to the number concentration of pre-existing particles with marine origin. A prescribed particle number concentration of 200 cm^{-3} was added to the lowest marine boundary layer, and exponentially decreased with altitude by

$$N (\text{cm}^{-3}) = 200 e^{-2(1-\sigma)} \quad \text{for } \sigma \geq 0.15$$

$$= 36.5 e^{-15(.15-\sigma)} \quad \text{for } \sigma < 0.15$$

where $\sigma = p/p_s$, and p_s is the surface pressure. These particles simulate a possible source of marine background particles such as sea salt. The simulated indirect forcing is then reduced by about 34% globally. These reductions are mainly over the ocean and are as large as 50% locally. This indicates that the global radiation balance is sensitive to the change in cloud drop concentration over the ocean. Therefore, the large extent of marine stratus and stratocumulus clouds may play important role in determining the net change in reflected solar radiation.

5. DROP EFFECTIVE RADIUS COMPARISON

Han et al. (1994) developed a scheme to retrieve cloud particle effective radii (r_e) in liquid water clouds using International Satellite Cloud Climatology Project

(ISCCP) data. Here, we compare their survey with our predictions to validate the accuracy of our cloud drop parameterization and to determine whether our treatment of cloud processes is reasonable (see Fig. 5). The general features of Figs. 5a and 5b are quite similar, although small discrepancies do exist. However, since the uncertainty in the retrieved data reported by Han et al. is about 1 - 2 μm , we conclude that the simulated cloud drop effective radii are in good agreement with those retrieved from satellite data and the effect of anthropogenic sulfate on cloud drop effective radii is more pronounced in the mid-latitude continents of the Northern Hemisphere (Fig. 5c).

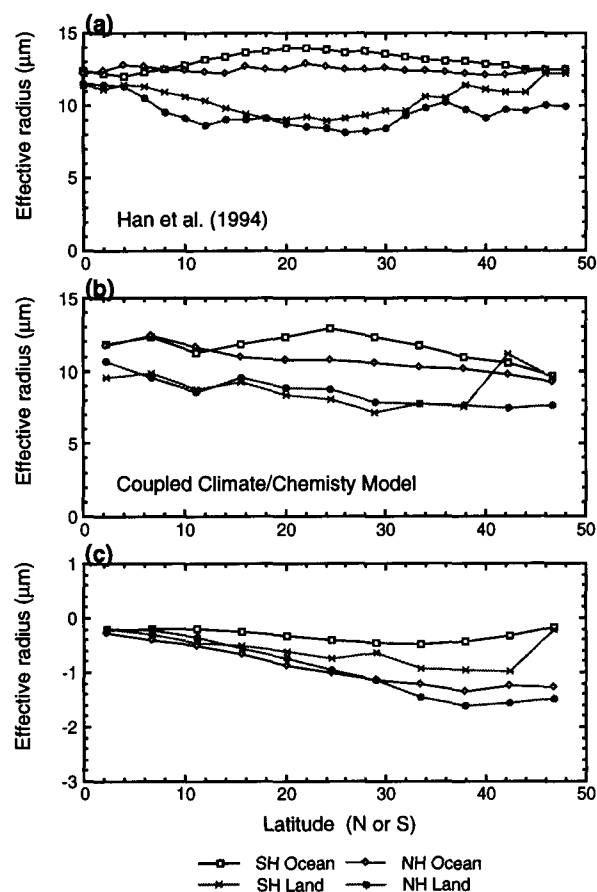


Fig. 5. (a) Annual, zonal mean cloud drop effective radii from NOAA-9 for 1987. (b) Same as (a) but simulated by the coupled climate/chemistry model using the internal mixing approach with an assumed aqueous conversion fraction of 75%. (c) Simulated changes in effective radii due to anthropogenic sulfate.

6. DISCUSSION AND CONCLUSIONS

Our estimate for indirect anthropogenic sulfate radiative forcing ranges from -0.6 to -1.6 Wm^{-2} and it reduces to -0.4 to -1.1 Wm^{-2} if a prescribed marine background particle number concentration is applied over the ocean. Uncertainty in the magnitude of the calculated indirect forcing may arise from the assumed

“pre-existing particle size distribution” which must result from a variety of processes and sources involving the entire suite of aerosol types (Penner et al., 1994). The prescribed pre-existing size distribution was chosen to represent an “average” particle distribution where no anthropogenic sulfate was present. Although it may remain questionable whether the pre-existing distribution is independent of anthropogenic sulfate and can be treated separately, we believe that the approaches used in this study provide a reasonable estimate of the possible range of forcing by the indirect effect.

7. ACKNOWLEDGMENTS

This work was performed under the auspices of the U. S. Department of Energy by the Lawrence Livermore National Laboratory under contract No. W-7405-ENG-48. We are grateful for the partial funding of this work by NASA under interagency agreement NAGW-1287 and by DOE under the Quantitative Links Program and Atmospheric Radiation Measurement (ARM) Program.

8. REFERENCES

- Charlson, R.J., S.E. Schwartz, J. M. Hales, R.D. Cess, J.A. Coakely Jr., J.E. Hansen, and D.J. Hofmann, Climate forcing by anthropogenic aerosols, *Science*, **255**, 423-430, 1992.
- Chuang, C.C., J.E. Penner, and L.L. Edwards, Nucleation scavenging of smoke particles and simulated drop size distributions over large biomass fires, *J. Atmos. Sci.*, **49**, 1264-1275, 1992.
- Chuang, C.C., and J.E. Penner, Effects of anthropogenic sulfate on cloud drop nucleation and optical properties, *Tellus*, **47B**, 566-577, 1995.
- Han, Q., W.B. Rossow, and A.A. Lacis, Near-global survey of effective droplet radii in liquid water clouds using ISCCP data, *J. Climate*, **7**, 465-497, 1994.
- Langner, J., and H. Rodhe, A global three-dimensional model of the tropospheric sulfur cycle, *J. Atmos. Chem.*, **13**, 225-263, 1991.
- Milford, J.B., and C.I. Davidson, The sizes of particulate sulfate and nitrate in the atmosphere - A review, *JAPCA*, **37**, 125-134, 1987.
- Penner, J.E., C.S. Atherton, and T.E. Graedel, Global emissions and models of photochemically active compounds, *Global Atmospheric-Biospheric Chemistry*, R. G. Prinn (Ed.), Plenum Press, New York, 223-247, 1994.
- Spiro, P.A., D.J. Jacob, and J.A. Logan, Global inventory of sulfur emissions with $1^\circ \times 1^\circ$ resolution, *J. Geophys. Res.*, **97**, 6023-6036, 1992.